Research Article | Vol. 30, No. 18 / 29 Aug 2022 / *Optics Express* 32074

Optics EXPRESS

Complementary dispersive mirror pair produced in one coating run based on desired non-uniformity

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Abstract: We report a novel one-coating-run method for producing an octave-spanning complementary dispersive mirror (DM) pair. The anti-phase group delay dispersion (GDD) oscillations are realized by two mirrors of the DM pair due to the certain thickness difference. Both mirrors are deposited within a single coating run enabled by the non-uniformity of the ion beam sputtering coating plant, which is obtained by tuning the distance between the source target and coating substrates. Since the DM pair is produced in a single deposition run, the GDD performance is more robust against deposition errors than that of the conventional complementary DM pair, in which two separated coating runs are necessary. Moreover, the new DM pair is compatible for both laser polarizations under the same angle of incidence, which could effectively reduce the difficulties of alignment for their implementation in laser systems than the double angle DM pair. The new DM pair is successfully applied to compress pulses from a Ti: Sapphire laser system down to 4.26 fs in pulse duration.

1. Introduction

Attosecond pulses offer tools to study electron dynamics in atoms [1], solids [2] and molecules [3]. The attosecond pulse generation relies on ultra-broadband few-cycle laser pulses in the visible/near infrared range [4,5]. Few-cycle laser pulses require perfect group delay dispersion (GDD) compensation over one octave spectral range, which makes the ultra-broadband dispersive mirror with the property of precise phase control [6–8] (DM, also known as chirped mirror [9]) the best way to compensate the dispersion over such a wide spectral range.

The undesired GDD oscillations, however, originating from impedance mismatching between the mirror and the incident medium have always accompanied DMs. A DM with large GDD oscillations could broaden the pulse duration and introduce satellite pulses, which will severely impede the pulse quality. Moreover, the larger the spectral bandwidth of the DM, the stronger the GDD oscillations. Several single mirror methods [10–13] have been proposed to suppress the GDD oscillations in the late 90s and the early 21st century. These methods, however, are not ideal for a broad one-octave spanning DM.

Wedge DMs [8], complementary DM pairs [6,14,15] and double angle DM pairs [7,16] are the most effective approaches to suppress undesired GDD oscillations over one octave. The wedge DM is a single mirror approach and composed of three parts, including an AR coating, a wedged layer and a multilayer stack. The wedged layer introduces high frequency GDD oscillations which are spectrally shifted along the gradient of the wedge. The wedge DM is able to provide low GDD

#467664 https://doi.org/10.1364/OE.467664
Journal © 2022 Received 12 Jun 2022; revised 16 Jul 2022; accepted 17 Jul 2022; published 17 Aug 2022

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oscillation with an odd number of reflections. However, the production of this mirror is very complicated. It needs three different coating runs to coat the three parts. Any deposition errors in the three coating runs could significantly increase the residual GDD oscillations. Moreover, the total thickness of the wedge DM is more than 20 μm, which can result in severe stress issues, such as delamination and fracture [17]. On the other hand, the two mirror-pair methods utilize two mirrors with anti-phase GDD oscillations to realize a smooth GDD. Considering the complex structure of wedge DMs, complementary DM pairs and double angle DM pairs are still the most widely-used few-cycle dispersion compensators so far [6,7,14,15,16–21], though each of them has their own limitations. For the complementary DM pair, two coating runs are necessary to produce the two different designs, which must be perfectly matched. The precision of the total thickness should be better than 0.1% from run to run. Higher deposition rate deviations will affect the spectral matching and result in an increased amplitude of the residual GDD oscillations. Therefore, producing a complementary DM pair in two coating runs is much more difficult than producing a DM in a single coating run. In contrast, the double angle DM pair consisting of two identical mirrors, which work at two different incident angles, can be produced in one coating run. Thus, the spectral matching is not a problem for a double angle DM pair. The disadvantage of the double angle DM pair, however, is that they must be accurately aligned in order to meet the angles of incidence (AOI) of the coating design, which increase the difficulty and time for the alignment. Moreover, small deviations of the AOI could lead to amplified GDD oscillations. Another disadvantage is that the GDD oscillations will be enhanced significantly for the other polarization, which makes it only work for the polarization specified in the design. In order to overcome these disadvantages, new mirror pair design and fabrication methods are highly desirable.

In [22], we proposed a quasi-complementary DM pair method to suppress the GDD oscillations of high dispersive mirrors with a relatively narrow working bandwidth. This method was realized by the spectral shift between the two mirrors caused by the inherent and constant non-uniformity as well as the post-deposition treatment. However, this method is not flexible due to the almost constant spectral shift. It also could not apply to an one-octave spanning DM pair, because the number of GDD oscillation increases a lot as the bandwidth increases and the GDD oscillations at different wavelength caused by post-deposition treatment might be different, which results in an imperfect match of the anti-phase GDD oscillation. Therefore, in this paper, a novel one-octave complementary DM pair based on the adjustable non-uniformity of the ion beam sputtering (IBS) coating process only is designed, produced and characterized. The DM pair consisting of two mirrors with a slightly different thickness, where the GDD oscillations are in anti-phase, could be produced within one coating run by using the non-uniformity effect. The non-uniformity is known to be the variation of physical thickness within the deposition area. The non-uniformity of IBS is mainly induced by the angular distribution of the sputtered particles. The distance between the target and substrates is a major parameter to adjust this distribution. Thus, the desired non-uniformity can be obtained by changing the distance between the sputtered target and substrates. The new complementary DM pair produced in one coating run overcomes several disadvantages of the conventional complementary DM pair, including two necessary coating runs and the spectral matching problem. Moreover, it works at the same AOI (5°), which has a lower sensitivity to the incident angle compared to double angle DM pair. Therefore, much less time is needed to align these new mirrors. The design approach of the new complementary DM pair is presented in section 2. The production process and characterization results of the DM pair is demonstrated in section 3. We draw our conclusions in section 4.

2. Design of the one octave complementary DM pair

The octave-spanning complementary DM pair is composed of two mirrors with a slightly different thickness. The anti-phase oscillations of the GDD are achieved by the spectral shift between the two mirrors due to the thickness differences. The physical thicknesses of two mirrors are proportional to each other, which means the thickness of the second one is related to the first one with a certain factor. This proportion could be realized by the non-uniformity effect in the coating chamber.

The DM pair was designed by the special multi-coating function included in the OptiLayer software [23]. In order to cover a wide-range from 500 nm to 1050 nm, Nb_2O_5 and SiO_2, which have a high refractive index ratio, were chosen as the layer materials. Fused silica (Suprasil, Heraeus) was used as the substrate. The optical constants of the substrate and layer materials are specified by the Cauchy formula:

$$n(\lambda) = n_\infty + A/\lambda^2 + B/\lambda^4 \tag{1}$$

where λ is in units of micrometers and the coefficients of n_∞, A and B are presented in Table 1.

Table 1. Cauchy formula coefficients for the layer materials and substrates.

	n_∞	A(μm^2)	B(μm^4)	n (0.8 μm)
SiO_2	1.486272	-3.996783E-3	5.8433165E-4	1.48769
Nb_2O_5	2.179779	0.032789	1.9913311E-3	2.23598
Suprasil	1.443268	0.004060	6.9481764E-6	1.44962

Both mirrors were designed to work at an incident angle of 5°. The thickness difference proportion between the two mirrors, (T_1-T_2)/T_1, was set to vary from 1.9% to 1%, where T_1 and T_2 is the total thickness of mirror 1 and mirror 2, respectively. The desired value can be realized in the production by changing the distance between sputtered target and substrate. The optimal proportion could be obtained during the minimization of the merit function. The designed mirror reflectance target was set to 100%. We choose to optimize the group delay (GD) with the special floating constants method [24], which allows us to shift the GD values up and down to get the best design without changing the amount of GDD. The GD difference between 1050 nm and 500 nm is about 82 fs. The designed GDD target, approximately -30 fs^2, at wavelength of 800 nm can be derived from the GD target. This mirror pair is optimized to compensate the dispersion of 1 m air and 1 mm fused silica with two reflections. The $GD(\omega)$ can be integrated from the $GDD(\omega)$ with an additional arbitrary constant C:

$$GD(\omega) = \int_{\omega_0}^{\omega_1} GDD(\omega)d\omega + C \tag{2}$$

The needle optimization [25] and gradual evolution [26] techniques were used to minimize the merit function to synthesize the design. The combined merit functions are described as follows:

$$MF(X,C)_\Sigma^2 = \sum_{j=1}^{500}\left(\frac{R_\Sigma(X,\lambda_j) - R(\lambda_j)}{\Delta R_j}\right)^2 + \sum_{j=1}^{500}\left(\frac{GD_\Sigma(X,\lambda_j) - GD(\lambda_j)}{\Delta GD_j}\right)^2 \tag{3}$$

where $\{\lambda_j\}$ are evenly distributed wavelength points in the spectral range from 500 nm to 1050 nm; $\{X\}$ includes $\{X^1\}$ and $\{X^2\}$, which are the vectors of layer thickness of the first mirror and the second mirror of the DM pair; R_Σ is the geometric mean of R_1 and R_2, which are the reflectance of the first and the second mirror, while GD_Σ is the arithmetic mean of GD_1 and GD_2, which are the GD of the first and the second mirror; $R(\lambda_j)$ and $GD(\lambda_j)$ are the reflectance and GD target, where $R(\lambda_j)$ equals 100%, $GD(\lambda_j)$ equals $GD(\omega)$ in Eq. (2) and therefore includes the constant C; ΔR and ΔGD are tolerances of reflectance and GD.

During the optimization, carefully adjusting the tolerances of the reflectance and GD is necessary. A one octave-spanning complementary DM pair design was synthesized as a result. The layer thicknesses and theoretical curves are shown in Fig. 1 and Fig. 2(a), respectively. One can see the slight thickness difference between the first and second mirror from Fig. 1. The total thickness of the first and second mirror is 9080.39 nm and 8916.94 nm, respectively. Due to this thickness difference, the GDD oscillations of the two mirrors are exactly in anti-phase, resulting in significantly suppressed GDD oscillations in the effective GDD, which is the arithmetic mean of the GDDs of the two mirrors, as shown in Fig. 2(a). The theoretical GDD of both mirrors oscillates from -820 fs^2 to 800fs^2, while the effective GDD oscillations of the DM pair are in the range of –140fs^2 to 70 fs^2, which is approximately one order of magnitude lower than GDD oscillations of single mirror. The designed results show the one-octave coverage of the DM pair between 500 nm and 1050 nm. The first and second mirror were labeled PC70-1 and PC70-2, respectively. The final thickness difference proportion of PC70-2 to PC70-1 is 1.8%. The proportion is achievable from the non-uniformity in the production.

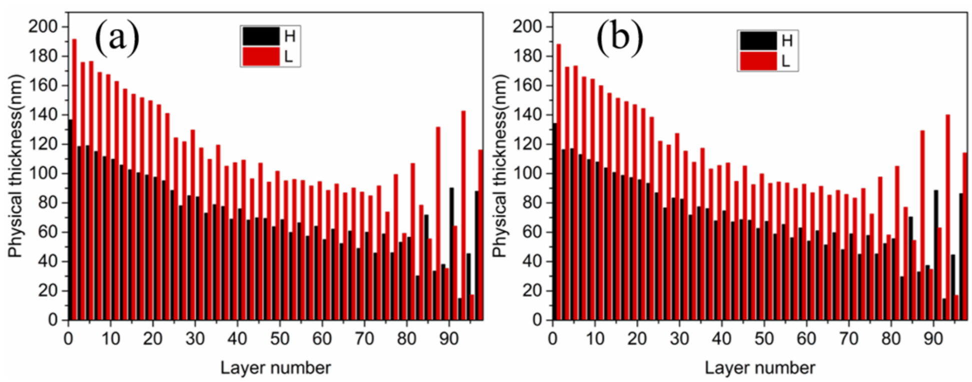

Fig. 1. Layer thickness structure of the two mirrors. (a) PC70-1, (b) PC70-2. Black and red bars represent the Nb_2O_5 (H) and SiO_2 (L) materials. The slight thickness difference corresponding to 1.8% between the two mirrors can be seen.

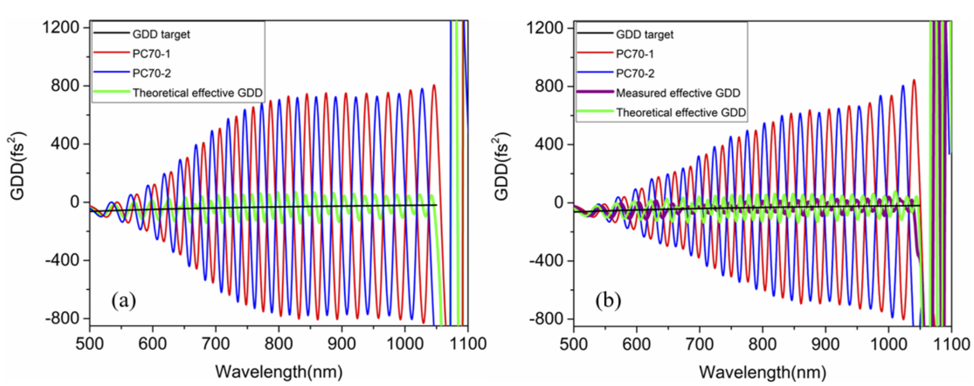

Fig. 2. Theoretical and measured GDD characteristic of the one octave DM. (a) Theoretical GDD characteristic, (b) Measured characteristic. Red and blue curves represent the first and second mirror named as PC70-1 and PC70-2. Green and black represent the theoretical effective GDD and GDD target. Violet curve shows the measured effective GDD.

3. Production and characterization of the one octave complementary DM pair

For ion beam sputtering the angular emission distribution for sputtered atoms from the sputtered target is often described as an under-cosine or heart shape distribution [27]. Due to such a distribution, the desired non-uniformity could be obtained by changing the distance between sputtered target and substrate fixture, which in our case is to adjust the substrate fixture height. In order to get 1.8% non-uniformity, we produced single Nb_2O_5 and SiO_2 monolayer and Quarter-wave multilayer coatings with different substrate fixture heights. During the multilayer process, changing the substrate height for Nb_2O_5 and SiO_2 materials can be easily done with the IBS control software. After the termination of one layer, the shutter will be closed and the substrate fixture will move to the correct height for the next layer. The transmittance spectra of these samples were measured by a spectrophotometer, which is presented in Fig. 3. The measured spectra were used for reverse engineering and calculating the non-uniformity, performed in the Optichar software [23]. Finally, the optimal substrate fixture heights, which are -35 mm for Nb_2O_5 and -50 mm for SiO_2 material respectively, were obtained to realize 1.8% non-uniformity between the third ring and fifth ring in the substrate fixture. The substrate fixture is made of different rings represented as the distance from the center of the fixture to each ring.

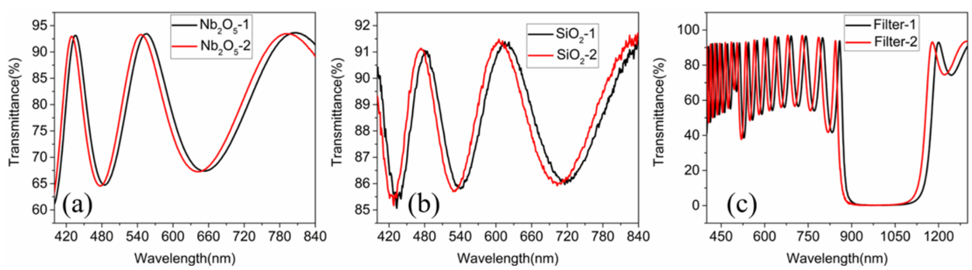

Fig. 3. Measured transmittance spectra of single Nb_2O_5, SiO_2 monolayer and a multilayer filter located at two different substrate fixture rings. Black and red curves correspond to the third and fifth ring, respectively.

After the desired non-uniformity was obtained, we started to deposit the DM pair. Two 1-inch Suprasil substrates were put at the two specified positions. The DM pair was produced with an ion beam sputtering machine from Cutting Edge Coatings GmbH (Hannover, Germany). The ion beam is extracted from an Argon gas plasma by a three-grid multi-aperture extraction system. A cryogenic pump evacuates the coating chamber to 1×10^{-7} mbar before deposition. The pressure during the coating process is about 5×10^{-4} mbar. The deposition rates are approximately 0.1 nm/s and 0.13 nm/s for Nb_2O_5 and SiO_2, respectively, and remain stable throughout the coating process, therefore, the layer thickness can be effectively controlled by the coating time of each layer. Meanwhile, the coating machine is also equipped with a broadband optical monitoring system, which allows us to monitor the in-situ transmittance.

The samples located in the third and fifth ring in the substrate fixture correspond to PC70-1 and PC70-2, respectively. The transmittance of both samples was measured by a Perkin-Elmer Lambda 950 spectrophotometer at normal incidence, and was plotted in Fig. 4. Excellent agreement between the measured and theoretical data of both mirrors can be observed from Fig. 4. Moreover, the spectral shift of the measured transmittance in the near infrared region (1050 nm to 1200 nm) between two mirrors is well reproduced as compared the theoretical results, which proves that the desired non-uniformity was realized with the coating parameters. The GDD was measured at the angle of incidence (AOI) of 5° and p-polarization by a home-built white light interferometer [28]. The measured GDD compared to the theoretical values is presented in Fig. 2(b). One can see that the measured GDD of PC70-1 and PC70-2 are in anti-phase and cancel each other, which results in much lower effective GDD oscillations. The measured GDD oscillates between -800fs^2 to 800fs^2, while the measured effective GDD oscillations of the DM pair are in the range of -120 fs^2 to 40 fs^2. The non-uniformity on the DM sample may lead to the imperfect matching of GDD oscillation between PC70-1 and PC70-2 and increase the effective GDD oscillation. However, with less than 0.1% non-uniformity on the DM sample the effective GDD oscillation of the DM pair is estimated to only increase a few percent. Moreover, the measured effective GDD fits well to the GDD target. The measured GDD curves have verified the realization of the complementary one-octave spanning DM pair, as can be seen in Fig. 2(b).

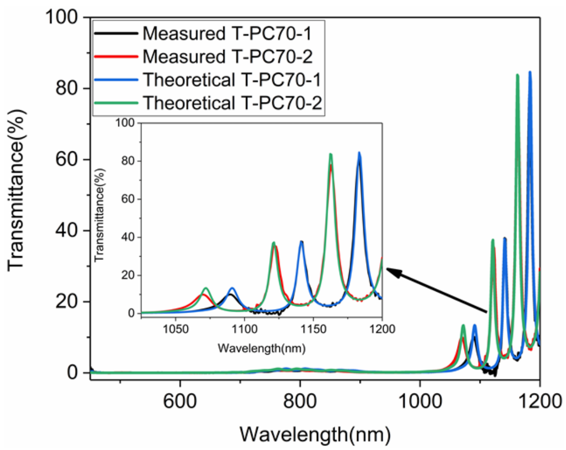

Fig. 4. Comparison of measured transmittance and theoretical transmittance of the two mirrors. Black and red curves represent the measured transmittance, while blue and green curves correspond to the theoretical transmittance.

In order to study the sensitivity of the new DM pair to the polarization and AOI, the GDDs were measured several times with different settings. The comparison of measured GDDs at both p- and s-polarization is shown in Fig. 5(a), while the comparison of measured GDDs at the incidence angles of 3°, 5° and 7° at p-polarization is presented in Fig. 5(b). As shown in Fig. 5(a), the measured GDDs are almost identical for both polarizations, which shows that the new complementary DM pair is robust against polarization changes. Moreover, Fig. 5(b) shows that the measured GDDs at an AOI of 3° and 7° are almost the same with a few nm spectral shift, verifying that the new complementary DM pair is insensitive to the AOI.

To estimate the compression effect of the new complementary DM pair for ultrashort pulse, we simulated the pulse envelopes reflected from the DM pair under different conditions. During the simulations, we assume that the input pulse is a Fourier transform limited super-Gaussian pulse with a pulse duration of 3.47 fs and a spectrum from 500 to 1050 nm, which is the working range of these mirrors. The input pulse was chirped by sending through an imaginary black box which has 10 times the amount of the target dispersion with opposite sign. Then the pulses were re-compressed by the DM pair or a single DM (PC70-1) with 10 bounces. We first simulated the pulse envelopes after 10 bounces on the DM pair at both p and s-polarizations, which are shown in Fig. 6. As can be seen from Fig. 6, the pulse envelopes and pulse durations after 10 bounces on the DM pair (blue and red curves) were almost the same to the input pulse for both polarizations. In contrast, the pulse envelope reflected from the single mirror of PC70-1 suffered significant changes, as the green curve in Fig. 6 shows. Huge losses of about 80% were observed due to the appearance of pre-and post-pulses. Moreover, the pulse duration was broadened to 6.76 fs. These simulations prove the excellent compression capability of the DM pair.

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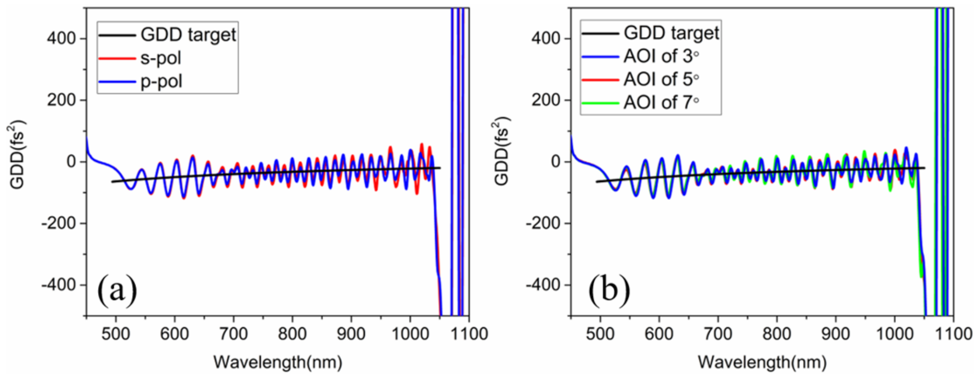

Fig. 5. Measured GDDs of the DM pair compared to the GDD target. (a) p and s-polarization, (b) angle of incidence of 3°, 5° and 7°.

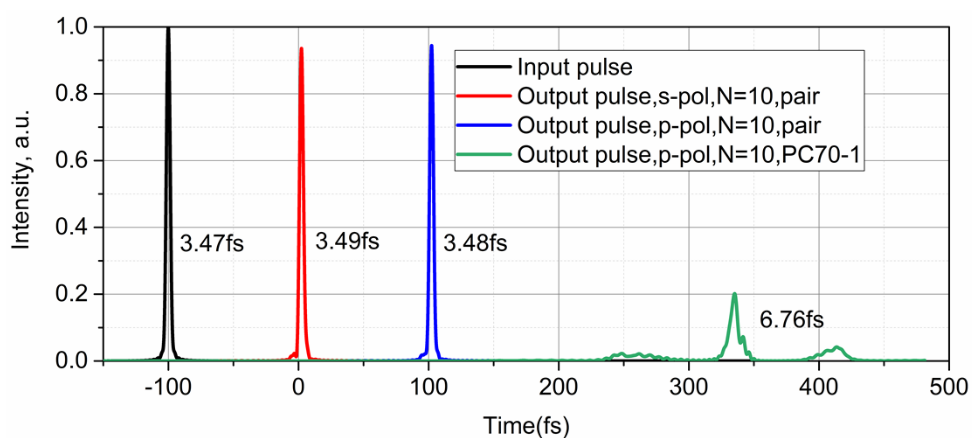

Fig. 6. Simulated input and output pulses under different conditions. Black curve represents the Fourier transform limited input pulse, red and blue curves correspond to output pulses compressed by the DM pair with 10 reflections at s-polarization and p-polarization, respectively. Green curve represents the pulse compressed by PC70-1with 10 reflections at p-polarization.

The one octave complementary DM pair was also tested experimentally with few-cycle laser pulses. The ultrashort pulses are generated by sending the output of a 10 kHz Ti:sapphire chirped pulse amplification (CPA) system through an Argon filled hollow-core fiber. The spectrally broadened pulses were then compressed by six pairs of the fabricated complementary DM. Moreover, to compensate the third order dispersion (TOD) a 2 mm thick ADP crystal was placed in the beam. The few-cycle pulses after the DM compressor were characterized with the dispersion-scan (D-scan) technique [29]. The measured D-scan map is presented in Fig. 7(a). The reconstructed spectral phase and temporal intensity profile are shown in Fig. 7(b) and Fig. 7(c), respectively with a reconstruction error of 0.18%. The pulse duration was retrieved to be 4.26 fs and the transform-limited duration is around 3.26 fs. Some residual TOD in the pulses after the DM compression give rise to a satellite structure at around -10 fs in the temporal intensity profile, shown in Fig. 7(c). The TOD of the pulses can also be seen as the tilt in the D-scan map. The residual higher order dispersion (e.g. TOD) can be further compensated by adding additional ADP crystal and DM pairs. However, experimental result already confirms the GDD compression capability of our newly developed complementary DM pairs.

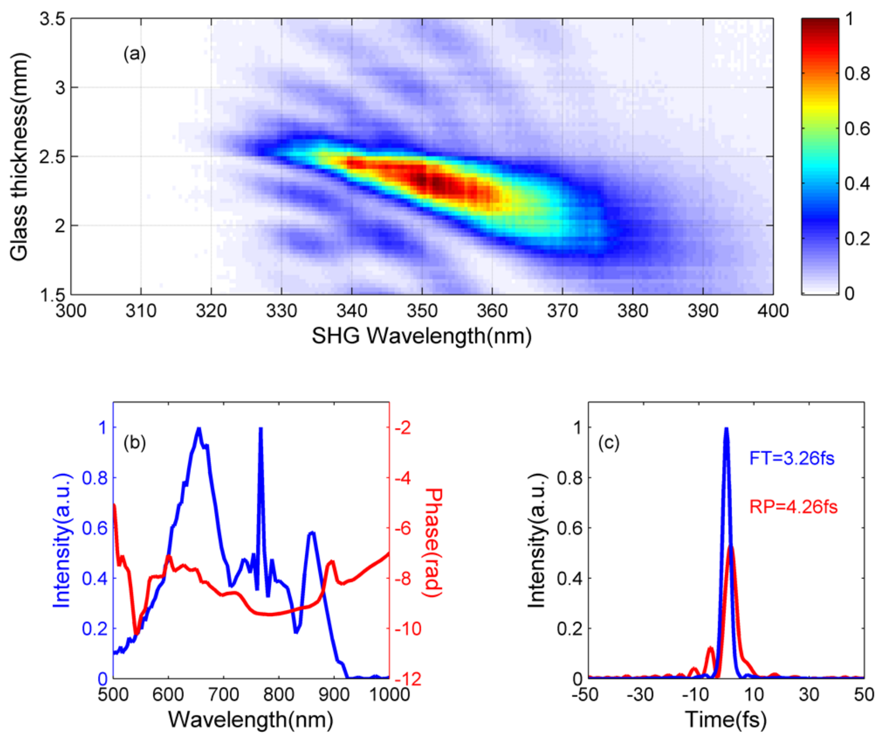

Fig. 7. (a) Experimental D-scan trace of a few-cycle pulse; (b) Measured spectrum (blue curve) and retrieved spectral phase (red curve); (c) Temporal intensity envelope of Fourier Transform limited pulse (blue curve) and retrieved pulse (red curve).

4. Conclusions

A new complementary DM pair which is able to provide -30 fs^2 GDD at 800 nm per bounce and compensate the dispersion of 1 m air and 1 mm fused silica over one octave has been successfully designed, produced and characterized. The DM pair composed of two mirrors with a slightly different thickness, where the GDD oscillations are just in anti-phase, is realized within one coating run based on the adjustable non-uniformity during the IBS coating process. With the complementary DM pair, we are able to compress pulses down to 4.26 fs in an actual ultrashort laser system.

Compared to the conventional complementary DM pair, the main advantage of this method is that the DM pair is produced in a single deposition run, which leads to a higher stability against deposition errors and less production cost and time. Furthermore, the new DM pair is used at the same incident angle and able to work at both polarizations making it much easier to be implemented in the laser system than the double angle DM pair.

Funding. Max-Planck-Gesellschaft; Munich Centre for Advanced Photonics.

Acknowledgments. The authors thank Prof. Ferenc Krausz for his valuable discussion and permanent support of this work. W. Li acknowledges support by the CSC program, and the Max Planck Society via the IMPRS for Advanced Photon Science. Z. Wang is grateful for support by the Alexander von Humboldt foundation. M.F. Kling is grateful for a Max Planck Fellowship from the Max Planck Society.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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